

OTIC FILE COPY

MISCELLANEOUS PAPER GL-87-27



A REVIEW OF RADON EMANATION AND MOBILIZATION IN MINERALS AND ROCKS

b

Christopher P. Cameron

Department of Geology University of Southern Mississippi Hattiesburg, Mississippi 39406



September 1987 Final Report

Approved For Public Release, Distribution Unlimited



Prepared for DEPARTMENT OF THE ARMY US Army Corps of Engineers Washington, DC 20314-1000

Monitored by Geotechnical Laboratory
US Army Engineer Waterways Experiment Station
PO Box 631, Vicksburg, Mississippi 39180-0631

87 11 5

010

When this report is no longer needed return it to the originator

The findings in this report are not to be construed as an official Department of the Army position unless so designated by other authorized documents

The contents of this report are not to be used for advertising publication, or promotional purposes. Citation of trade names does not constitute an official endorsement or approval of the use of such commercia. The Easts

Unclassified

ECURI	TY CLASSI	FICATION OF	THIS PAGE

REPORT D	OCUMENTATIO	N PAGE			Form Approved OMB No. 0704-0188
1a REPORT SECURITY CLASSIFICATION Unclassified		16 RESTRICTIVE	MARKINGS		
28 SECURITY CLASSIFICATION AUTHORITY			AVAILABILITY OF		
26 DECLASSIFICATION / DOWNGRADING SCHEDU	E		oution unlim		156,
4 PERFORMING ORGANIZATION REPORT NUMBE	R(S)	5 MONITORING	ORGANIZATION R	EPORT NU	MBER(S)
		Miscel	laneous Pape	r GL-87	1-27
6a NAME OF PERFORMING ORGANIZATION Department of Geology, Univer-	6b OFFICE SYMBOL (If applicable)	7a NAME OF M USAEWES	ONITORING ORGAN	NIZATION	
sity of Southern Mississippi		Geotecl	nnical Labor	atory	
6c. ADDRESS (City, State, and ZIP Code)		76 ADDRESS (Ci	ty, State, and ZIP (ode)	
Hattiesburg, MS 39406		PO Box Vicksb		80-0631	
Ba. NAME OF FUNDING/SPONSORING	86 OFFICE SYMBOL	9 PROCUREMEN	T INSTRUMENT ID	ENTIFICATI	ON NUMBER
ORGANIZATION US Army Corps of Engineers	(If applicable)				
8c. ADDRESS (City, State, and ZIP Code)		10 SOURCE OF	FUNDING NUMBER	s	
		PROGRAM ELEMENT NO	PROJECT NO	TASK NO	WORK UNIT ACCESSION NO
Washington, DC 20314-1000				1	
11 TITLE (Include Security Classification)			<u> </u>	<u> </u>	
A Review of Radon Emanation a	and Mobilization	in Minerals	and Rocks		,
12 PERSONAL AUTHOR(S) Cameron, Christopher P.	· · · · · · · · · · · · · · · · · · ·				
13a TYPE OF REPORT 13b TIME CO Final report FROM		14 DATE OF REPO		Day) 15	PAGE COUNT
16 SUPPLEMENTARY NOTATION	to	September	1987		.9
Available from National Techr VA 22161.	oical Informatio	on Service, S	5285 Port Ro	yal Roa 	d, Springfield,
17 COSATI CODES	18 SUBJECT TERMS (Radiation (L			identify b	y block number)
FIELD GROUP SUB-GROUP	Rocks (LC)	o) min	erals (LC)		
	Radon (LC)				
19 ABSTRACT (Continue on reverse if necessary Determination of the nat			and its var	iation	should receive
high priority during screening					
underground facilities. Rado					· · ·
rocks of the earth's crust.ar terms of human exposure to na					
excavations are limestones, g					
lents) all of which have low					
other factors often preclude lithologic settings, there is					
ation environments.					
Radon daughters are natu concentration may be anomalou					
				(Con	tinued)
20 DISTRIBUTION / AVAILABILITY OF ABSTRACT		21 ABSTRACT SE	CURITY CLASSIFICATION	ATION	
☐ UNCLASSIFIED/UNLIMITED ☐ SAME AS R	PT DTIC USERS	22b TELEPHONE	(Include Area Code		FICE SYMBOL
DD Form 1473. JUN 86	Previous editions are	absolete		C) A 55 (5) C	ATION OF THIS PAGE

Unclassified

COUNTY COCCUES INSCIONAL CONTROL

SECURITY CLASSIFICATION OF THIS PAGE

19. ABSTRACT (Continued).

those in granitic rocks. Anomalous accumulations of radioactive minerals will generate radon gas in above-background quantities. In deep unlined excavations, radon gas can contaminate the air as it is easily adsorbed onto dust particles, soot (from motors), and water droplets, as the gas emanates from rock faces, broken rock, faults, joints, fractures, and underground waters entering the excavation. Radon concentrations above atmospheric levels have been found in such protected places as underground low-level radiation counting facilities, which normally are constructed with heavy walls of low activity concrete. On the basis of laboratory evidence and field measurements, the mobilization of radon and its precursor isotope, radium, is often more extensive than originally predicted by earlier studies and conceptual thought; albeit, that the mechanisms controlling such mobilizations are, as yet, incompletely understood.

The geochemical behavior of daughter isotopes in the radioactive decay series of uranium and thorium is generally not well understood. It would be advantageous to know more about the emanating power of different types of uranium and thorium minerals particularly with respect to mineralology, grain size, porosity, and permeability. Considerable work is needed on ground-water geochemistry and the geochemical behavior of radon precursors in relation to anomalous concentrations of radon in the underground environment. The extent of radon mobility and transportation at depth by various convective mechanisms including atmospheric and other "pumping" effects, with respect to large underground excavations, is speculative as are the details of the effect of subsurface temperature variations and zones of high heat flow. Efforts to develop better instrumentation and passive radon detection systems need continued support.

A significant number of scientists feel that too little is known about the long-term health and genetic effects of prolonged exposure to low-level radiation environments. Radon has been recognized as playing a central role in the low-level radiation environment. Numerous research programs have been undertaken to measure radon concentration in human habitations and dwellings and to determine the impact of variable radon concentrations on health safety.

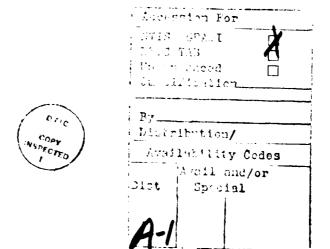
Unclassified

PREFACE

This Miscellaneous Paper is the result of a request from the Geotechnical Branch, Engineering and Construction Directorate, Office of the Chief of Engineers, to the Geotechnical Laboratory (GL), US Army Engineer Waterways Experiment Station (WES), to prepare a short report summarizing the major aspects of radon generation, emanation, and escape in crystalline rocks (particularly granites and their metamorphic equivalents).

The report was prepared during Fiscal Year 1987 by Dr. Christopher P. Cameron, Associate Professor of Geology, University of Southern Mississippi. General supervision was provided by Dr. D. C. Banks, Chief, Engineering Geology and Rock Mechanics Division, GL; and Dr. W. F. Marcuson III, Chief, GL.

COL Allen F. Grum, USA, was the previous Director of WES. COL Dwayne G. Lee, CE, is the present Commander and Director. Dr. Robert W. Whalin is Technical Director.



CONTENTS

	Page
PREFACE	1
INTRODUCTION	3
BackgroundObjectivesScope	3 4 4
BASIC PRINCIPLES	5
The Natural Radiation Environment	5 7 7 9
RADON EMANATION	10
Background Radon Emanation Radon Emanation Characteristics Radon Emanation and Temperature Diffusion of Radon through Granitoid Rock	10 10 12 12 22
MECHANISMS OF ESCAPE OF RADON FROM PRODUCTION SITES	24
Background Escape Mechanisms-Radiation Damage in Minerals Temperature Annealing Effects on Radon Escape	24 24 26
RADIOMETRIC ANOMALIES AND THEIR CAUSES	26
Background Radon Migration Common Causes of Radiometric Anomalies Temporal Variations in the Natura Radiation Environment Surface Variations	26 28 29 31 31 31
ENVIRONMENTAL PROTECTION AND HEALTH HAZARDS	32
Radiation Environments Sadon Detection and Monitoring Systems	32 33
RESEARCH DIRECTIONS	34
REFERENCES	36
BIBLIOGRAPHY	36
APPENDIX A. NOTATION	A 1

A REVIEW OF RADON EMANATION AND MOBILIZATION IN MINERALS AND ROCKS

INTRODUCTION

Background

- l. Increased US Army Corps of Engineers (CE) involvement in site characterization and construction of underground facilities in crystalline rock situated at considerable depths may require that some study and consideration be given to:
 - a. The possibility that anomalous accumulations of radioactive uranium and thorium minerals might be discovered during the excavation phase of the project; particularly if the facility is to be sited in ancient granites which make up substantial portions of the interior continental shield nucleii. For example, we know that major accumulations of uranium and thorium minerals (substantial tonnages with average grades in the 500 6000 ppm range) appear to occur preferentially near the unconformities separating (1), Archean granitoids and lower Proterozoic metasedimentary sequences and (2), intruded lower Proterozoic metasedimentary sequences and middle Proterozoic sequences. Some late Precambrian granites (especially some alaskites) have extensive pegmatitic phases which (in some cases) are rich in radioactive minerals.
 - b. Occupational risk and environmental protection with respect to possible anomalous occurrences of radioactive minerals which could provide increased dosages of radon gas (a possible cause of lung cancer) to personnel involved in the excavation, construction, and occupation of such facilities. It is pertinent to note that radon dosage in all habitations has become a topic of high interest and has been the subject of several recent meetings and short courses on a national level.
 - Methods and techniques of radon detection and monitoring including prediction of potential accumulations of radioactive minerals and daughter products during the exploration phase of site characterization.
 - d. Remedial and preventive methods to inhibit radon gas accumulation in confined underground spaces. These methods might be integrated with others employed to minimize occupational risk resulting from the need to blast, transport, crush, and hoist large tonnages of rock in large mined spaces, and for personnel to occupy those spaces. These operations create the need for fresh air ventilation to clear away dust, blasting gases, diesel

- engine emissions and other fumes, all of which can adsorb radon gases if the latter is present in appreciable amounts.
- 2. Radon is found everywhere on earth, in the atmosphere, hydrosphere, lithosphere, and biosphere. The radon emanation characteristics of rocks. soils, and minerals are of special interest to audiences interested in the way radon circulates in the lithosphere, the way this circulation affects the natural radiation environment in general, and environmental surveillance in particular. The concentration of airborne radioactivity can become high in the confined spaces of any underground excavation which encounters anomalous accumulations of radioactive minerals and/or daughter products of the natural uranium and thorium decay series. Epidemiological studies conducted over the past three decades by the Public Health Service in the United States have indicated that uranium miners have a higher incidence of lung cancer than the general population. These studies have also indicated that the radiation dose from the daughter products of Radon 222 (218 Po, 214 Pb, 214 Bi, 214 Po), as one of the causes of lung cancer. Hence the need for enhanced awareness with respect to the radiation environment of deep mined facilities; particularly in geological settings prone to anomalous accumulations of radioactive minerals.

Objectives

- 3. The objectives of this report are to:
 - a. Briefly summarize basic principles and hypotheses which describe radon emanation, mobilization, and escape from naturally occurring radioactive minerals of uranium and thorium.
 - \underline{b} . Outline the health hazards related in inhalation of excessive amounts of radon gas.
 - c. Describe those areas where knowledge of the low-level underground radiation is incomplete or missing altogether and applied research is indicated.

Scope

4. This report is the result of a request from the Geotechnical Branch, Engineering and Construction Directorate, Office of the Chief of Engineers (OCE), to prepare a report summarizing the major aspects of radon generation, emanation and escape in crystalline rocks (particularly granites and their metamorphic equivalents). The scope of this report will include a

discussion of the basic principles governing the naturally occurring radioactive decay of uranium and thorium, as well as brief comments concerning the controls on anomalous accumulations of the radioactive minerals. Radon emanation and escape mechanisms and the effect of temperature variations are discussed as are health hazards associated with inhalation of excessive amounts of radon. Common radon detection and monitoring systems are outlined. The report concludes with general recommendations regarding further studies and applied research.

BASIC PRINCIPLES

5. Radon (Rn), element 86 in the Periodic Table is the heaviest member of the rare gas group (Group 0). Like other members of this group, radon is an inert gas and thus does not enter into chemical combination with other elements in nature. There are three isotopes of radon: Rn-222, Rn-220 (sometimes called Thoron), and Rn-219 (sometimes called Actinon). These isotopes are daughter products of the three radioactive decay series Uranium-238 (U-238), Thorium-232 (Th-232), and Uranium-235 (U-235) respectively. Because U-235 has a low abundance in nature (0.7 percent of natural U) the contribution of Actinon to the natural radiation environment is minimal and will not be discussed further.

The Natural Radiation Environment

6. The majority of natural radiation from non-cosmic sources originates from the decay series outlined above and from the Potassium-40 decay series. These decay series are shown schematically in Figure 1 (from Saum and Link, 1969). The numbers between adjacent blocks are the half-lives of the isotopes in the decay series. For example, 4.5 billion years (Y) between uranium-238 and thorium-234 means that, in that time, half of the original uranium atoms will decay to thorium. Correspondingly, 3.82 days (D) between radon-222 and polonium-218 means that, in that time, half of the original radon atoms will decay to polonium. The horizontal connecting line indicates that this decay will result in alpha particle (ray) emission. Inclined lines indicate the emission of a beta particle (ray). Those isotopes which are surrounded by heavy lines are significant gamma-ray emitters and are therefore very

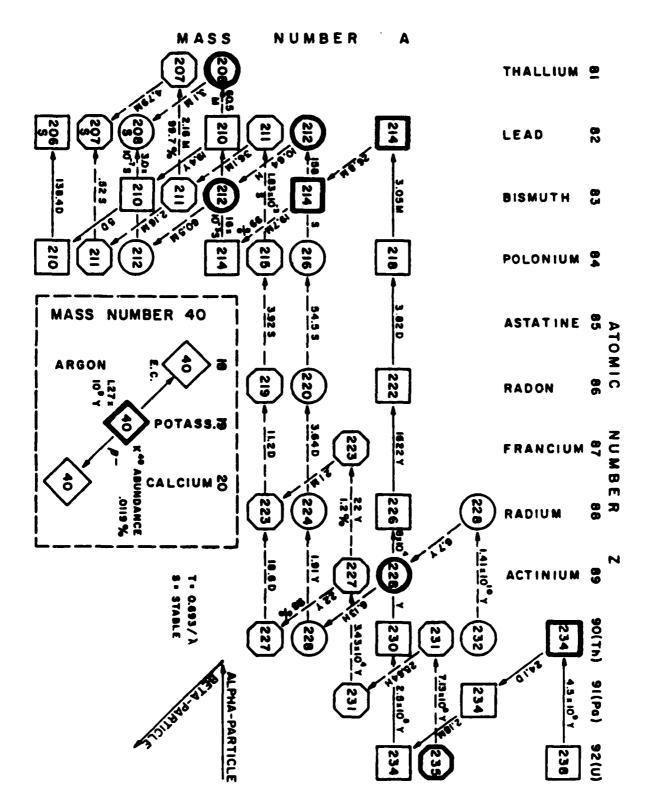


Figure 1. The natural radioactive decay series of Uranium-238, Thorium-234, and Potassium-40 (After Faul, 1954)

important contributors to the natural radiation background (Th-234, U-235, Ac-228, Pb-214, Pb-212, Bi-214, Pb-212, T1-208, and K-40).

Gamma Radiation

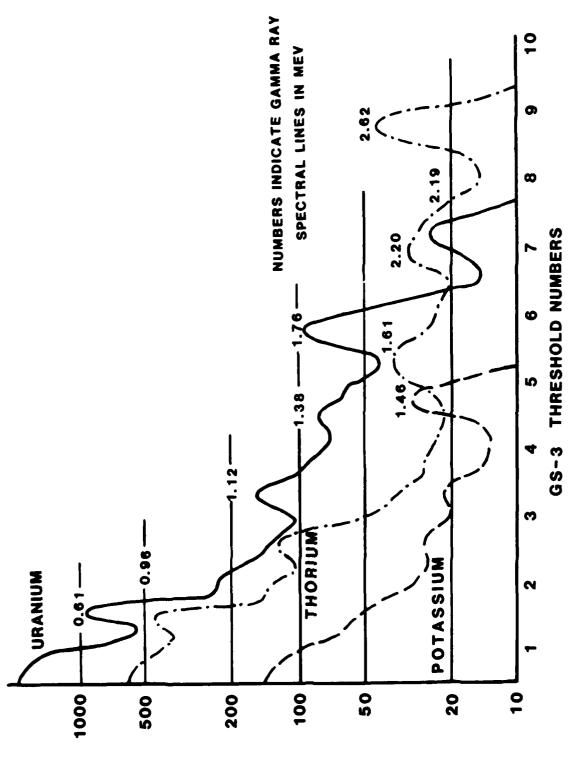
7. Gamma rays are quanta of energy which are very similar to x-rays in their velocity and nature. However, whereas x-rays originate in the electron shells of the atoms, gamma rays emanate from the nucleus. Gamma rays have the highest energy level of the radioactive decay emissions, can penetrate rock thicknesses of more than a foot, and can travel thousands of feet through air if the source is sufficiently concentrated and of significant mass. Figure 2 is a typical gamma ray spectrogram for the decay series of thorium, uranium, and potassium. Gamma ray detection systems which discriminate between radioisotopes of the three decay series on the basis of the threshold peaks (shown in Figure 2) are frequently employed in the exploration of uranium and mineral resources, geological mapping, in the characterization and documentation of the terrestrial gamma radiation environment, and in evaluating the impact of nuclear industries on the environment. These detection systems also integrate the gamma radiation to give a "total count". However, counters of this type should not be used to quantitatively estimate the activity of alpha and beta particles because alpha and beta emissions have much lower energy levels and often do not penetrate into the counting chamber(s) of the gamma ray instruments.

By defining the International Commission on Radiological Protection (ICRP) standard in terms of the dominant hazard, radon daughters, a certain amount of radiation exposure is accounted for automatically in terms of radiation dosages to underground workers. However, in some high-grade uranium deposits, the dominant hazard may be external exposure to gamma radiation. Since this radiation irradiates the entire body whole body limits must be used in terms of ICRP standards.

Alpha Radiation

8. The horizontal lines connecting adjacent blocks in Figure 1 indicate that the decay is by emission of an alpha particle. Alpha particles are composed of two protons and two neutrons. As they are expelled from the nucleus they rapidly strip electrons and are neutralized. When electrically stable,

RELATIVE COUNT RATE



SOLVE BASSASSE PARAGESSE ANNOUNCE PROGRAMME ACCORDANCE ANNOUNCE ANNOUNCE PROGRAMME PARAGES PROGRAMME PARAGES

Figure 2. Typical gamma ray spectrogram for potassium, uranium and thorium decay series

alpha particles become helium atoms. Because of their affinity for electrons alpha particles do not travel far and can be attenuated by thin layers of solid materials (e.g. a few sheets of paper). Despite the rapid attenuation of alpha radiation over short distances, the fact that radon is a gas and can be inhaled poses a health hazard (especially if dosages are excessive) because, in this case, the alpha ray attenuation will take place in the tissues of the lung.

Beta Radiation

9. Beta particles are equivalent to electrons and are produced by the decay of a neutron into a proton and a beta particle. Beta particles have no mass and exhibit relatively low energy levels. Beta particles can travel further and faster than alpha particles but like alpha particles are rapidly attenuated by a thin layer of solid material. Because of this alpha and beta particles are often referred to as "soft radiation".

Radon Precursors

10. As is indicated in Figure 1, both radon-222 and radon-220 (Thoron) are alpha emitters. As the decay series makes clear, radon should not considered without reference to the elements before it in the series. Of particular importance is radium-226 and radium-224. It must always be remembered that the distribution of radon in natural materials, rocks, soils, and waters, is related to, and reflective of, the distribution of radium. In some cases radium distribution may be reflective of the original uranium and/or thorium distribution but this is not always the case, especially in surface and near surface environments where parent - daughter isotope separations are common. The nature and extent of such separations in deep crystalline rock settings is a matter of some conjecture. However, results from deep drilling programs in the Granite Mountains (Wyoming), and, more recently, geothermal investigations, indicate that fluid transport and the leaching of metals (including uranium) along avenues of enhanced porosity and permeability occur widely and suggest that uranium - radium separation is probably more common than previously thought.

RADON EMANATION

Background

- 11. The phenomenon of radon escape from radioactive minerals was described in the early part of this century by Boltwood (1905, 1907, 1908) in his study of the origin of radium. He introduced the term "emanating power" to describe the loss of emanation from radioactive minerals although the radionuclide radon was not recognized as such in these early works. Modern literature pertaining to radon emanation and escape mechanisms includes the work by P. M. Barretto (1971) which is contained in his M.A. thesis, "Radon-222 Emanation from Rocks, Soils and Lunar Dust", (Rice University). Barretto's work is also summarized in "Development of Remote Methods for Obtaining Soil Information and Location of Construction Materials Using Gamma Ray Signatures for Project THEMIS". This five volume series of reports was completed in the period 1970 - 1972 by the Department of Geology, Rice University, under the direction of Dr. J. A. S. Adams, et al. for the USAE Waterways Experiment Station (WES) under Contract No. DACA 39-69-C-0048. Most of what follows regarding radon emanation and escape mechanisms, including Tables 1, 2, 3 and Figures 3 - 7, was compiled from their reports.*
- 12. Barretto (1971) notes that although literature treating radon emanation and loss from radioactive minerals (particularly those rich in uranium) is relatively extensive, until the development of highly sensitive alphascintillators combined with photomultiplier techniques, the emanation data on common rocks and soils were very few. As is indicated in the data contained in the aforementioned Tables and Figures this situation has changed considerably over the past two decades.

Radon Emanation

13. For every atom of radon produced there are two alpha-emitting daughters produced a short time thereafter (equilibrium is attained within four hours). Thus, radon emanation and escape in soils and rocks can be determined relatively quickly in properly equipped laboratories using cores

^{*} Notations for these tables and figures are shown in Appendix A.

and other soil and rock samples. Radon escape has been detected from almost all rocks and soils including those with very low parent uranium or thorium contents (e.g. basalts, gabbros, and even ultramafics).

- 14. The 3.82 day half-life of radon-222 produced from radium-226 in rocks is sufficiently long that extensive radon migration of radon gas can occur provided the radon can escape from its production site. How radon escape rates through earth materials vary for different lithologies and soils and what the escape mechanisms are for the escape of radon from its production site are important questions for those interested in determining the natural radiation environment and its temporal variation. It is pertinent to note that, along with preferential leaching of U-238, U-234 (Thurber 1962, Rosholt et al. 1963, Doe and Newell 1965), or Ra-226 (in chloride and carbonate charged waters) and the precipitation of Th-230 and Ra-226 by sulfate bearing waters, radon emanation constitutes one of the major causes of disequilibrium in the uranium series.
- 15. In the surface and shallow subsurface, mechanisms which control radon soil gas migration include "pumping" by variations in atmospheric pressure, wind, temperature, and moisture. In deeper geological environments (where radon is assumed to be dissolved in formation waters) the mechanisms of radon transport (and the migration of its precursor isotope, radium) are more speculative. Rapid groundwater flow along structural avenues (perhaps influenced by convection), as well as transport by the sweeping effect of other gases (CO₂, CH₄, H₂S) which migrate in the subsurface, are mechanisms that have been proposed to explain radon anomalies over deeply buried sources; but clearly more research is need in this area particularly on a site specific level.
- 16. Although the mechanisms are not well understood, laboratory and field evidence suggests that radon can migrate at rates well above those predicted by its low diffusivity in solid rock below the water table. As described in detail below (Diffusion of Radon through Solid Rock) radon concentrations above atmospheric levels have been measured in such protected places as low level radiation counting facilities, sited below ground level, and constructed with heavy wall of low activity concrete!

Radon Emanation Characteristics

17. The physical characteristics of Ra-222 emanation, as determined by the extensive laboratory studies of Barretto (1971) and Adams et al. (1970 - 72), are summarized in Tables 1, 2, and 3 and are shown graphically in Figures 3, 4, and 5. From these data we can draw the following general conclusions:

PERSONAL PROPERTY STANDARD CARAMAGE STREETINGS

- a. Rocks are generally poor radon emanators (relative to soils and some minerals) with the exception of granitoids which can experience radon loss of more than 10 percent.
- b. Basic igneous rocks (basalts, gabbros, serpentinites) and some sedimentary rocks (quartzose sandstones, orthoquartzites, limestones) have very low emanations. The low emanations are caused by low parent element concentrations.
- c. Radon release from sedimentary rocks ranges widely, however, if combined, the total far exceeds that for the total igneous rock radon release into the atmosphere. As is illustrated in Figures 3 and 4, conglomerates dominate radon escape activity.
- d. Soils (Table 2) display a broad range in emanation rates and radon loss but escape-to-production ratios are generally significantly higher than those experienced by most rocks and minerals. The wide variation may be caused by the substantial range of physical state and mineralogy shown by soils.
- e. Results from weathered samples (E-827 and INK-G in Table 1) indicate that weathering is an important factor in increasing radon emanation. In this case the increase in radon emanation is probably a function of enhanced effective porosity which results from the breakdown of rock matrix and mineral degradation during weathering.
- f. Although some common accessory minerals (Table 3) have high uranium concentrations and consequently high emanation rates their escape-to-production percentages are low both in value and range. These results show that the percentage of radon which escapes from a mineral is not necessarily correlated at all with the uranium concentration. It is believed that radon escape is more largely dependent upon the stability of the mineral structure and its crystallinity.

Radon Emanation and Temperature

18. The influence of temperature on the rate of radon emanation was investigated in experiments conducted by Barretto (1971), and others of the Department of Geology at Rice University on samples from the Graniteville Granite. Their results are illustrated in Figure 6 and show that for

Table 1
Radon Emanation Characteristics

KKSKISSO SESSOON SESSOON OSESSOON SESSOON SESSOON

Rocks

Sample	Rock Type	eU (ppm)	$^{222}_{\text{Rn Activity}}$ pci. h ⁻¹ gm ⁻¹ x10 ⁻⁴	Emanation Rate Rn atoms h gm	Fscape-to- Production (%)
VEB-437	Gnetss	9.0	2.26	4.0	14.0 ± 0.76
VCS-34	Gnetss	0.4	0.14	0.2	1.0 ± 0.26
I-275A*	Qtz-monz-gneiss	11.9	7.27	12.8	2.4 ± 0.08
E-833*	()tz-monz. orthogneiss	1.7	5.41	9.5	12.6 ± 1.58
E-829*	Otz-dior, paragneiss	4.0	8.26	14.5	7.9 ± 0.82
E-830*	Granodiorite	2.3	9.81	17.3	16.9 ± 1.9
E-827*	Granodiorite	2.0	20.20	35.6	40.0 ± 3.1
VCS-25	Granodiorite	1.9	1.84	3.1	3.9 ± 0.16
VFS-276	Granodiorite	1.6	4.92	8.7	12.2 ± 1.28
VFS-297	Otz. diorite	9.0	1.07	1.9	4.7 ± 0.87
VTR-1571	Otz. diorite	3.2	4.76	8.7	6.1 ± 0.97
E-832*	Otz. monzonite	3.0	6.98	12.3	+i
VEB-14	Granite	6.0	1.56	2.7	+1
VEB-365	Alaskite	1.0	3.69	6.5	+1
VEB-640	Granite	3.0	24.80	43.7	+1
VCS-26	Pegmatite	7.8	8.48	14.9	+i
¥292-I	Pegmatite	14.4	11.00	19.4	+i
¥1/2-I	Porph. granite	14.6	46.30	81.6	12.5 ± 0.28
E-828*	Granite	6.1	62.20	109.5	+1
E-831*	Granite	1.9	4.53	8.0	9.4 ± 0.92
CON-C	Granite	12.2	11.80	20.9	3.8 ± 0.39
MILF-G#	Granite	0.7	0.86	1.5	4.8 ± 0.47

From P. M. Barretto, (1971) and J. A. S. Adams et al. (1972).

(Sheet 1 of 3)

Table 1 (Continued)

Sample	Rock Type	eU (ppm)	$^{222}_{Rn\ Activity}_{pci.\ h^{-1}\ gm^{-1}x10^{-4}}$	Emanation Rate Rn atoms h gm	Escape-to- Production (%)
GRAN-G#	Granite	9.6	22.10	39.0	+1
CHEL-G#	Granite	8.5	16.80	34.0	7.8 ± 0.63
GR-G	Granite	2,3	4.28		
FM-100	Granite	2.0	4.29	7.5	+1
INK-G	Granite (W)	3.0	12.40	22.6	+ı ℃
VM-274	Dacite	7.6	16.30	28.8	6.9 ± 8.1
E-823*	Syenite	5.3	12.40	21.9)+ S
NIM-1*	Lugaurite	14.7	57,30	0.66	15.4 ± 0.74
MBT-57	Basalt	0.4	0.28	0.5	+1 cc
MBT-3	Basalt	1.3	0.84	1.5	+1
IPS-193	Basalt	2.3	4.52	7.9	7.7 ± 1.0
V-1 ★	Diabase	• 05	1.19	2.1	+I
1-274*	Gabbro	4.7	4.25	7.5	3.6 ± 0.58
AAE-256	Volc. tuff	55.0	23.2	41.0	1.7 ± 0.12
I-268*	Conglomerate	9.4	2.71	8.4	+1
1-269*	Conglomerate	0.6	23.40	41.2	10.4 ± 0.48
I-263*	Metacomglomerate	12.3	80.90	142.5	26.0 ± 0.91
SP-PR	Serpentinite	0.5	0.10	0.2	1.0 ± 0.63
¥212-1	Quartzite	7.2	19.2	33,7	+1
GC-27	Quartzite	0.2	2.7	0.5	5.3 ± 0.49
3C - 22	Quartzite	9.0	3.0	0.0	1.9 ± 0.19

(Continued)

(Sheet 2 of 3)

Table 1 (Concluded)

CONTRACTOR SECURISE S

Sample	Rock Type	(mdd)	pci. h gm x10-4	Rn atoms h gm	(2)
OKL-FR	Sandstone	3.5	10.5	18.6	11.9 ± 0.94
AAC-218	Sandstone	5.1	4.18	7.3	3.2 ± 0.40
AAE-309	Sandstone	34.0	45.5	80.2	5.3 ± 0.22
AAF-374	Sandstone	3.4	07.9	11.2	7.4 ± 0.71
OKL-ST	Red Silt	2, 1	4.31	7.6	8.1 ± 1.02
E-825*	Dol. subarkose	8,3	43.20	20.7	5.6 ± 0.53
AUST-L	Limestone	1.9	99.0	1.3	1.6 ± 0.42
EL-AB-A	Asphal. limestone	6.4	2.24	4.2	0+
EL-AB-0	orl. Limestone	2.6	0.40	0.7	0+
SPL-L	Limestone	1.7	0.74	1.3	1.7 ± 0.56
EL-DOC-L	I.imestone	5.5	3.11	5.5	0 +I
3-25	Silt shale	3.2	62.3	10.9	7.7 ± 0.41
CC-3	Shale	1.2	8.10	14.2	2.67± 0.16
EP-7	Laterite (Fe)	24.2	17.5	30.8	2.86± 0.19
RM-2	Laterite	8.9	5.7	6.6	2.52± 0.15
LIP-60	Volc. glass	6.5	1.3	2.1	0.51± 0.04

All samples were crushed and sieved < 60 > 115 mesh unless indicated. \star = grain sizes < 200 mesh # = NBS rock standard & = Weathered

(Sheet 3 of 3)

Table 2
Radon Emanation Characteristics
Soils

Sample	Soil Type	el.	222 Rn Activity pci. h ⁻¹ gm ⁻¹ ×10 ⁻⁴	Emanation Rate Kn atoms h gm	Escape-to- Production (7)
JBR-1	Calcareous	3.1	26.1	97	33
MER-1	: :	9.0	3.4	ِ ج ا	3 (
CC-1	=	2.9	7.07		ν. V
LSI-15	Elluvium	2.8	25.6	r 	38
LSI-25	=	3.3	16.8	<i>5</i> €	Ü,
S-0K-2	Alluvium	2.3	7.6		<u></u>
INK-25	Granitic	7.7	29.0	15	9 7
INK-35	=	3.7	35.0	6.1	1.4
S-0K-3	Sandy (Dune)	1.3	3.1	S	σ
S-0K-4	" (Loess)	3.3	16.5	δ.΄	0
S-0K-7	Clayev	2.9	13.8	۶.	<u>0</u>
TSI-D		2.8	25.6	57	~
SPT-P	=	0.	36.0	5 }	30
POT-5	Volcanic	3.5	43.8	r.	o -1
EL-DOC-1	Residual (Calc.)	5.0	86.0	150	5.
EL-DOC-2		7.5	95.2	167	7,7
S-BAUX	(Ba	1.2	ະຄ. ກ•	1.5	30
J-BAUX	=	17.4	59.4	105	71
LGN-P	Lignitic	3.4	31.5	95	36
LM-3-GL S-410-2	Glauconitic	0.8	0.7	39	m m

From P. M. Barretto, (1971) and J. A. S. Adams, (1972).

Radon Emanation Characteristics Table 3

Minerals

Mineral	e((ppm)	222 Rn Activity pci. hour - 1 m - 1	Emanation Rate Rn atoms h l m	Escape-to- Production (%)
Zircon (20)*	416-2660	$0.4-30 \times 10^{-2}$	140-5220	0.2-4.8
$Zircon(W)^{\#}(1)$	405	1.2×10^{-1}	2180	12.1
Sphene (15)	29100	$0.8-68 \times 10^{-4}$	4-742	0.2-4.7
Biotite (2)	311	2.3×10^{-3}	36–33	2.8-6.6
Monazite (1)	627	1.5×10^{-2}	39	0.2
Xenotime (1)	6028	3.5×10^{-4}	260	0.09
Apatite (1)	17	3.3 x 10 ⁻⁴	y	0.8
Allanite (1)	ر ، 0	8.2×10^{-5}	5.8	0.3
Magnetite (Ti) (1)	0.8	$1-18 \times 10^{-4}$	1.5	0.4
Glauconite (?)	1 2 2		1–39	3.0

(20)* number of samples analyzed. (W) weathered

From Barretto, (1971) and 1. A. S. Adams (1972).

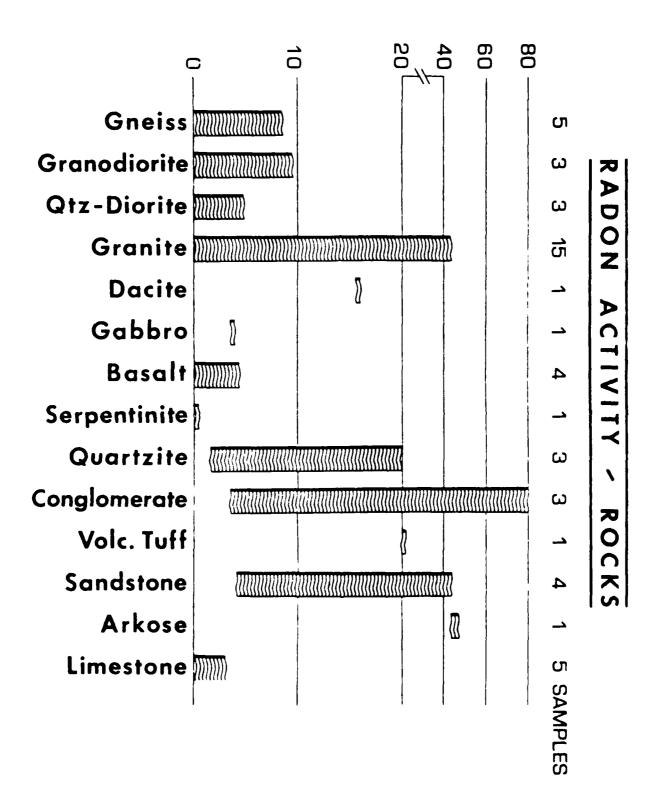


Figure 3. (From P. M. Barretto, 1971)

RADON 222 ESCAPE TO PRODUCTION RATIO (%)

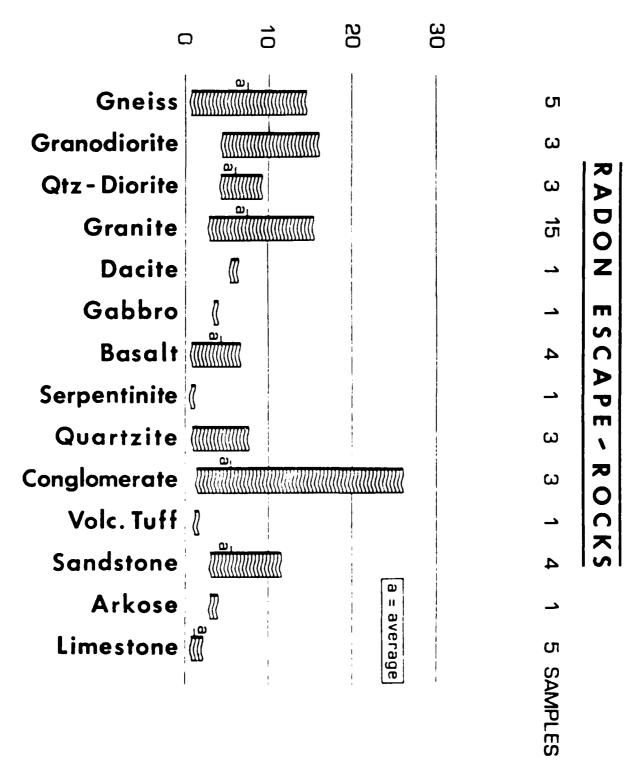


Figure 4. (From P. M. Barretto, 1971)

RADON 222 ESCAPE TO PRODUCTION RATIO (%)

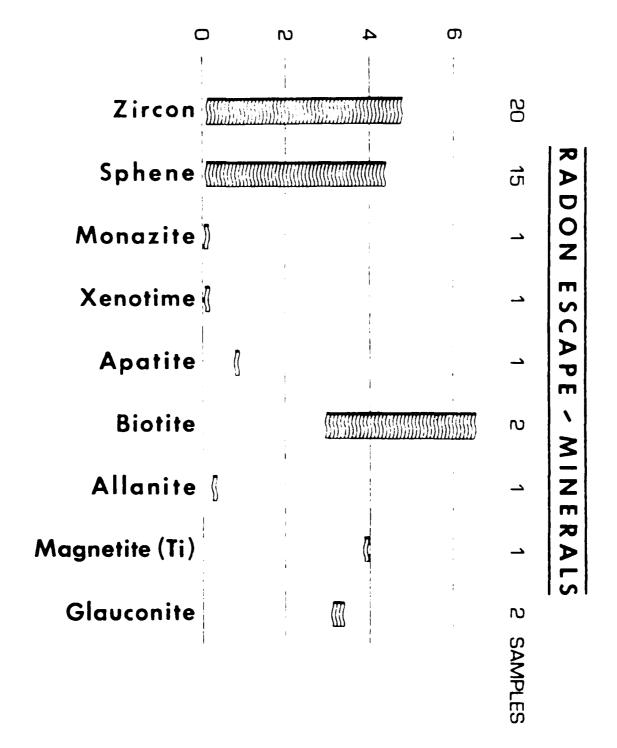


Figure 5. (From P. M. Barretto, 1971)

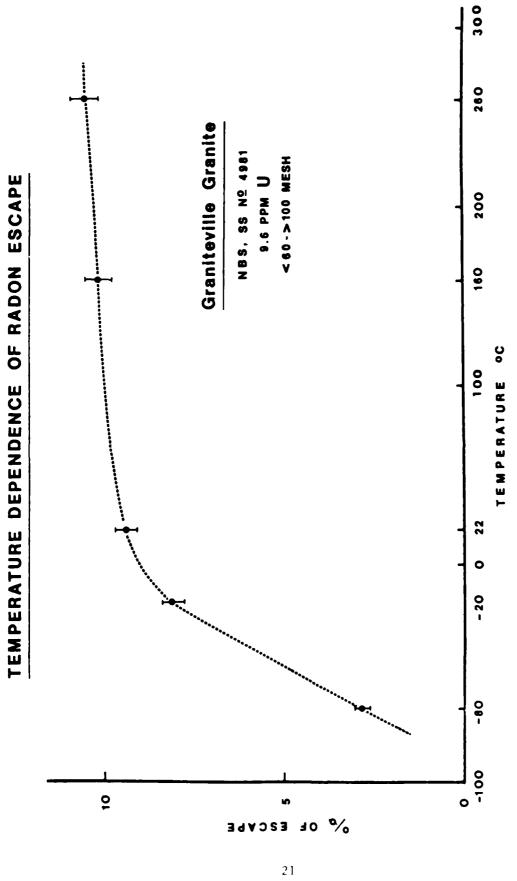


Figure 6. (From P. M. Barretto, 1971)

temperatures in the range 25 - 265 degrees (C) only a slight increase in the percentage of radon escape is observed. At low temperatures radon release is drastically reduced; by 13.5 percent at -20 degrees (C) and by 70 percent at -80 degrees (C). These results agree with Barretto's (1971) work on radon behavior at low temperatures using highly emanating soil samples.

Diffusion of Radon through Granitoid Rock

19. As has been previously indicated, the rate of radon migration through rocks has been observed to be higher than that predicted by its low diffusivity. Guedalia et al. (1970) studied radon diffusion in soils and produced a laboratory model which predicted a radon mobilization of Radon-222 (half-life 3.825 days) of 360-420 m. Barretto (1971) considered this depth "unrealistically large"; and then proceeded to produce experimental results (using crystalline rocks) which told much the same story and left him at a loss to understand what appears to be a rapid radon mobilization mechanism operating in granites. The following excerpt is from Barretto's (1971) observations and his published results on radon diffusion experiments which used granite cores from the Conway Granite batholith (New Hampshire).

"Although these experimental observations indicate rapid diffusion of radon through substantial thicknesses of solid fresh granite, it is difficult to understand the mechanism especially considering the accepted low permeability of granite and absence of pressure changes. Previous reports of this are not found in the literature. It is interesting, however, to recall that radon concentrations above atmospheric levels have been found in such protected places as low level radiation counting facilities, below ground level, which usually are constructed with heavy walls of low activity concrete. In these cases, the radon migration into the underground facilities has been attributed to the presence of fractures in the concrete, even if they are not apparent. Due to the lack of independent data which would or would not support the above laboratory observations, the interpretations or derived consequences will not be discussed further. The diffusion of helium through glass is well known and used industrially. By analogy, the radon results may indicate that although granite and glass are generally impermeable to most fluids, the glass has spaces that permit the ready diffusion of helium and the granite may have spaces that allow the radon to diffuse rather rapidly (see for example Brace et al. 1972)."

20. As part of "Granite Resurgence of Water Level" (GROWL) LeGrand (1986) developed a hydrogeological framework hosting radon emissions from indoor air, as well as radon and radium emissions from well water, to provide a conceptual model to account for high concentrations of these isotopes under certain water conditions in fractured crystalline rocks. In this model the required conditions for accretion of radon and radium (which can then dislodge and enter tightly insulated habitations or well water) are (1), granite with normal (2-4 ppm) or above normal amounts of uranium; (2), an interconnecting fracture pattern; (3), a mantle of relatively impermeable material over the granite; and (4), repeated cycles of a fluctuating water table in the fracture zone. In this model, as radium and radon are mobilized by the fluctuating water table and by variations in atmospheric pressure which produce a "pumping" effect. It is effectively trapped by the impermeable zone where it accretes. This entrapment results in a slight pressure flow to least resistant outlets in the vicinity.

(Author's Note: The above model of radon transport mechanism may be useful in explaining the accumulation of anomalous radon gas in "sealed" underground excavations. Almost all such facilities usually have some sort of "least resistant" outlet or inlet as an integral part of their structure.)

Radon anomalies over sources at significant depths in sedimentary rock sequences are also documented in the literature. Strong (2 - 3 times background radon anomalies have been recorded over Starks Salt Dome in Calcasieu Parish (Louisiana) where the depth to cap rock is in excess of 1200 feet (370 meters) and to salt 1925 feet (589 meters). These anomalies are apparently controlled by marginal ring and interior radial faults; structures which enable ground water movement and barometric changes (as well, as, perhaps, the "sweeping" effect of the migration of other gases) to mobilize radium and radon from these depths, (Gabelman, 1972). A similar situation prevails over the Chacahoula Salt Dome in Lafourche Parish where the depth to cap rock is 932 feet (285 meters) and to salt 1367 feet (418 meters). In other cases structural controls to enhance the mobilization of radium and radon from substantial depths are not evident. Gingrich (1975) reports radon anomalies over a relatively low-grade uranium deposit in the Grants Mineral Belt (New Mexico) which is at a depth of 360 feet and overlain by a Dakota (Cretaceous) sandstone sequence which contains interhedded shale stringers and

- a thin coal seam. In this case the presumed source, the uranium deposit, is located above the water table.
- 22. In summary, it can be stated on the basis of laboratory evidence and field measurements made during uranium exploration programs that the mobilization of radon and its precursor isotope, radium, is often more extensive than originally predicted by earlier studies albeit that the mechanisms of such mobilizations are, as yet, incompletely understood.

MECHANISMS OF ESCAPE OF RADON FROM PRODUCTION SITES

Background

23. The escape of radon from radioactive minerals was noted by Boltwood (1905, 1907, 1908) in his study of the origin of radium. He also noted that the "emanating power" of a mineral was considerably reduced by heating. Lind and Whittemore (1914), found "emanating powers" in the range 16 - 50 percent when they investigated the radium - uranium ratio in the mineral carnotite. Holmes (1948) was the first to relate radon emanation to discordance in the uranium - lead system, an observation which held the interest of geochronologists for over a decade (Giletti and Kulp, 1954; Kulp et al. 1954; Horne and Davison, 1955; Eckelman and Kulp, 1956) until they concluded that radon leakage played an insignificant role in explaining the isotopic ages of the samples they analyzed. Instead lead loss events, which are more suitable to detection and are the major effects in the uranium-lead system discordance, became emphasized; and the radon emanation mechanism relegated to much lower importance in geochronological research (Barretto, 1971). Fortunately, research (in the United States) by General Electric Laboratories and Rice University Department of Geology during the period 1960 - 1975 produced important findings and a new understanding in radon escape mechanisms.

Escape Mechanisms - Radiation Damage in Minerals

24. Radiation damage to mineral crystal lattices produced by spontaneous fission fragments, alpha particles, and recoil nuclei provides the means of radon escape (Barretto, 1971). It is important to note that micas and other potassium minerals are the sites of production of radiogenic argon and

xenon. These lighter, noble gases do not escape from their production sites and for this reason are useful in geochronological studies. Yet radon does escape and mechanisms other than the one outlined above (thermal diffusion, leaching, diffusion through microfissures) fail to account for the observed radon loss in laboratory experiments. Barretto (op. cit.) and Fleisher et al. (1964, 1965, 1966) describe the following important aspects of radiation damage in minerals as well as the radon escape mechanism:

- Part of the radiation damage in radioactive minerals or in minerals which host radioactive elements, that produced by fission tracks, can be observed under a microscope after the damaged portions of the mineral structure are selectively etched by chemical treatment.
- <u>b</u>. The fission tracks are randomly oriented, frequently intersect one another, and their density or numbers per unit volume varies according to uranium distribution in the mineral.
- Much more abundant are radiation damage effects due to alphaparticles and recoil nuclei. These are more difficult to observe directly however, because of their smaller size, and hence more difficult to evaluate.
- d. If there is no radium-226 leaching then the radon production site must be at or very near the uranium lattice site. The radon generated is dislocated from its production site by the nuclear recoil energy (4.78 MeV for the alpha-particle and 90 KeV for the Radon-222 atom). Because many other alpha disintegrations took place in the uranium decay chain before reaching radon-222 (see Figure 1) this region of the crystal must be highly damaged. A fraction of the radon moved by the recoil can diffuse to the grain surface by utilizing all possible pathways connected to the damaged zone.
- e. The particular emanation rate of a mineral will be a function of several independent variables including concentration of radioactive elements, age, and crystal structure. Also the site of this radioactivity, whether inside or outside the lattice, in solid solution, occlusion or inclusion, or fracture filling, is important as it will control the distribution and pattern of the radiation damage.
- f. Radon leakage from a mineral starts at a low baseline level and will increase with time as the crystal undergoes damage from the decay of uranium and/or thorium and their daughters. Radon escape should theoretically reach a maximum when the mineral crystal structure is completely destroyed.
- g. All minerals do not respond in the same manner to radiation damage. For example, zircon inclusions in feldspars fail to show the pleochroic haloes so common when zircon is hosted in biotite; suggesting an equilibrium between damage rate and recovery which varies between mineral species. Some minerals

- structures(s) are readily damaged by low alpha-dosages while others fail to show any detectable effect.
- h. Thorite, phyrochlore, euxenite, and zircon (all of which are relatively abundant in some granitoid terranes) are examples of uranium rich minerals in which varying degrees of metamictazation (an advanced stage of radiation damage) have been found. However, autenite, metatorbenite, sphene, and xenotime are reported as not being subject to metamictization despite their high uranium concentrations.
- <u>i</u>. Temperature plays the most important role in annealing the radiation damage to mineral crystal structure.

Temperature Annealing Effects on Radon Escape

25. Barretto (1971) and Fleisher et al. (1965) conducted extensive experiments on the annealing effects of temperature, pressure, healing time, plastic deformation, and ionizing radiation. There is little doubt that temperature plays a dominant role in annealing radiation damage in crystals. Figure 7 illustrates the variation of radon loss with temperature increase for various mineral species and for granite. Radon loss is not significant below 100 degrees (C) but, for granite, becomes important in the range 100 - 600 degrees (C).

RADIOMETRIC ANOMALIES AND THEIR CAUSES

Background

26. The potential increase in demand for fissionable elements to fuel nuclear reactors sparked intensive, if sporadic, uranium exploration activities during the 1950-1980 period. These activities resulted in extensive literature which deals with radiometric anomalies and their causes. Allied literature treating methods of radiometric data analysis and anomaly discrimination and interpretation are equally voluminous. It is beyond the scope of this report to discuss radiometric anomalies in other than outline form. The interested reader is referred to the extensive bibliographies published by the US Department of Energy and The International Atomic Energy Commission for more comprehensive treatment of this subject.

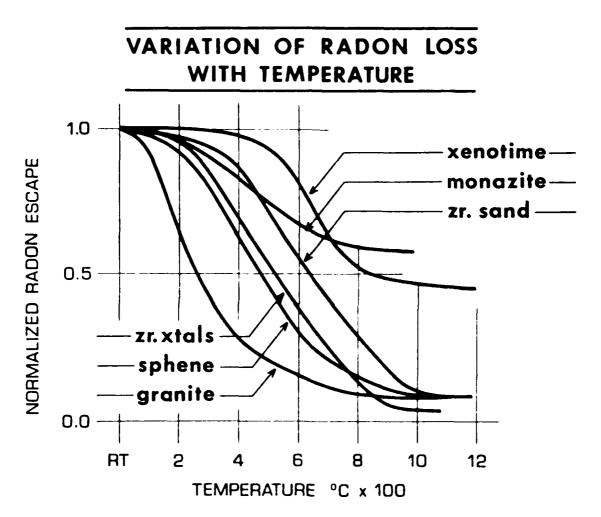


Figure 7. (From P. M. Barretto, 1971)

27. It is useful to define "radiometric anomaly" or "anomalous radio-activity". An enormous amount of time, energy, and money has been spent in attempting to establish what is meant by these terms by those in resource estimation, exploration and development, as well as by those involved with safeguarding health and the environment; and interpretations vary somewhat. "Anomalous radioactivity" is defined herein as a significant deviation from uniformity and regularity in radiometric quantities measured by the techniques and instrumentation developed to date. As such, radiometric anomalies are caused by the natural accumulation of decay series radioisotopes in quantities measurably above statistical background which are established by detailed surveys of the natural radiation background in the area or region of interest.

Radon Migration

- 28. Radon will migrate by both diffusion and transport in a fluid media (air or water). However, diffusion of radon-222 in a water-saturated porous media such as that pertaining in most subsurface geological environments is so small that diffusion alone cannot account for the radon anomalies which have been observed in the field. Clearly, a transport mechanism is responsible for the radon associated with most anomalies.
- 29. Several mechanisms have been observed to operate depending on whether the radioactive source is situated above or below the water table (e.g. atmospheric pumping, convection, sweep by other gases, high rates of fluid flow in faults and other structural avenues, and combinations of all of these).
- 30. Dispersion of, (for example), uranium and its radium daughter product from a deeper source can result in radium concentrations at the water table. This in turn results in an upward diffusion (or other transport) of radioactive soil gas (radon) which can reach the surface and be detected by both gamma-ray scintillometry and alpha-ray detection systems. (The amount of soil gas present on a given day is very much a function of atmospheric pressure; hence the use of the passive detection systems (e.g. alpha-cup detectors) which remain in place for a month or so and "mean-out" the atmospheric effect.)

31. It should be noted that mobilization and transportation of radon and its precursors at depths significantly below the water table is still, poorly understood.

Common Causes of Radiometric Anomalies

- 32. The following phenomena commonly act to produce radiometric anomalies:
 - a. Structural avenues of groundwater movement. Structural avenues of fluid migration such as faults, fracture zones, and regional joint sets, have long been noted for their often anomalous radiometric signatures. Recalling that radon has a very short half-life and that its diffusion rates are low, and that fluid flow rates typically range from 1.5 meters/day to 1.5 meters/ year in most geologic media, it is difficult to explain many radiometric anomalies which have their source at substantial depths. In some cases structural controls, perhaps aided by "sweep" of other gases (CO₂, H₂S, CH₄, etc.) must enable groundwater flow and barometric pressure to mobilize radium and radon from great depths. In other cases structural controls to support the mobilization of radioactive isotopes from substantial depths are not evident.
 - b. The role of groundwater geochemistry. The solubility of uranium (and the important radon precursor radium) is covariant with salinity. Studies which treat geothermal systems and hydrothermal minerals deposits suggest that convective transport of groundwater at depth in some crystalline terranes can yield a hydrothermal fluid with high salinity and acidity; particularly in areas of anomalously high heat flow. As salinity and acidity of the fluid increases so does its capacity to leach metals from the country rock. The result, as indicated in studies of geothermal systems and fluid inclusion investigations, is a brine. The degree to which the fluid is enriched in a particular metal is dependent on the abundance of the metal in the rock, and the geochemical factors which control rock-water interaction and its rate.

The above mechanism has been proposed to account for some hydrothermal mineral deposits in granitic terranes including uranium and thorium accumulations in veins and disseminations. This mechanism also accounts for anomalous amounts of radium and radon in some geothermal systems and hot springs. Again, structural features such as faults and fractures offer the optimum avenues of fluid migration in crystalline terranes.

c. Anomalous (including economic) concentrations of uranium and thorium. Parent elements can concentrate in anomalous amounts through a variety of well-recognized processes in sedimentary and crystalline rock settings. For example, some continental sediments host a tuffaceous volcanic and/or a siliceous

volcano-clastic component. Leaching of this component will enrich groundwater in uranium. Transported in groundwater as uranyl-dicarbonate, uranium may precipitate in reducing environments produced by: the metabolic activities of anaerobic bacteria feeding on organic debris in the sediments, carbonaceous debris or humic acid accumulations, the introduction of hydrogen sulfide gas associated with hydrocarbons at depth, and related phenomena which result in oxidation - reduction reactions along chemical fronts. These processes have acted to produce economic concentrations of uranium in continental sandstones in many parts of the world.

Other anomalous accumulations of uranium and thorium minerals in sedimentary rocks include the radioactive placer deposits hosted in Lower Proterozoic conglomerates in Canada, South Africa, and Brazil. Anomalous uranium concentrations in the calcretes of some arid terrains are not uncommon.

SEER PRICEOUS SUBSIDIOS BRIGHAN BRIGHT BRIGHT BRIGHT

In younger settings thorium and uranyl-thorium mineral accumulations can occur when thorium-rich heavy minerals form part of the sediment load in fluvial and nearshore marine depositional environments. Thorium-rich placer deposits are often the result. These produce total radiometric anomalies similar to those sourced by uranium concentrations. As is always the case when both uranium and thorium occur together, anomaly discrimination must be accomplished by spectral analysis or, if the thorium source material is right on the surface, by chemical methods. Modern gamma-ray spectrometers can usually resolve the ambiguity inherent in anomalies with a significant thoron contribution.

Uranium and thorium are lithophile elements and have an affinity for acidic igneous rocks, particularly granites and their extrusive equivalents. Those portions of granitic intrusions which have undergone "wet" hydrothermalism and the crystallization of pegmatitic differentiates are often enriched in uranium and thorium minerals. Uranium and thorium can concentrate to economic grades in such phases (e.g. Bancroft District, Canada; Rossing, Southwest Africa). Pegmatitic terranes are thus an obvious source of anomalous radium and radon.

d. Lithologic variations. The physical and chemical processes involved in the formation and later alteration of a given rock unit often involves the enrichment of parent and daughter isotopes with the results that the radiometric signature of the unit contrasts sharply with that of adjacent units. For example, the Chattanooga Shale Formation carries an anomalously high uranium background, as do many acidic plutonic and extrusive igneous rocks (and their metamorphic equivalents). Limestones generally have very low levels of radioactivity. The list of potential variations in radioactivity among rock types is extensive (see Figures 3 and 4). Fortunately these variations tend to have regional implications, and can usually be defined as background variations rather than local anomalies, particularly during follow-up surveys.

Temporal Variations in the Natural Radiation Environment

Surface variations

- 33. Temporal variations in the natural radiation environment (as measured on the earth's surface or in the atmosphere) are caused by variations in temperature, barometric pressure (which can produce a pumping effect), wind, moisture, and variations in the regional or local water table. These variations can take place over periods which range from hours to months and, in the case of water table variation, even years. More importantly, radon concentrations can vary by as much as a factor of 100 in a 24 hour period. Consequently, short term measurements of soil gas such as those done by traditional emanometer techniques are usually not reflective of average radon concentrations in a given area. However, a considerable amount of experimental evidence suggest that most temporal variations in radiometric background "mean-out" over a period of 21-30 days.
- 34. Because of the variability of weather conditions emanometer readings are usually not repeatable and the use of passive alpha-detectors for radon activity measurement is now generally recommended. By leaving the detector cups buried in shallow excavations for periods in excess of one month the detectors continuously accumulate the readings produced by varying radon soil gas concentrations and thus produce a reading indicative of long term average.

Subsurface variations

35. Variations in radon activity at depths significantly below the water table should be less and occur over a longer period of time than those in the shallow subsurface. Deep geological settings are generally water saturated except in those environments where liquid or gaseous hydrocarbon fluids are present or in areas of pronounced geothermal activity where boiling of fluids is possible and rapid vapor-liquid phase changes are likely. In deep water-saturated settings the parameters which so drastically affect radon gas variation in the near-surface environment (changes in barometric pressure, temperature, moisture, and wind) have minimal effect and variations should be minimal. However, other factors 'mentioned earlier) which control the mobilization and transportation of radon and its precursors may be impacted by the deep excavation and construction of large underground facilities. Continual monitoring of the background radiation in and near the facility site during

site characterization, construction, and facility occupation is indicated on the basis of present knowledge and understanding of the subsurface natural radiation environment.

ENVIRONMENTAL PROTECTION AND HEALTH HAZARDS

Radiation Environments

- 36. The concentration of airborne radioactivity can become high in the confined spaces of any underground excavation which encounters anomalous accumulations of radioactive minerals and/or daughter products of the natural uranium and thorium decay series. Epidemeological studies of conducted over the past three decades by the Public Health Service in the United States have indicated that uranium miners have a higher incidence of lung cancer than the general population. These studies have also indicated that the radiation dose from the daughter products of Radon 222 (218 Po, 214 Pb, 214 Bi, 214 Po), as one of the causes of lung cancer. However, the amount of radiation dose delivered to the lungs by inhaled radon is uncertain because the dose depends on many factors, such as the dustiness of the air in the excavation, the length of time the air has been in circulation, the breathing rate, mucus in the bronchial passages, and the physiology of the bronchial passages as affected by smoking or infection. The amount of radon entering an excavation is determined by the rate of emanation into the mine, the rate of removal by ventilation, and the radioactive decay of the gas.
- 37. Although the above concerns are legitimate levels of radon accumulation in deep excavations of interest to the CE should be orders of magnitude less than those experienced in most uranium ore mines or mines where uranium is recovered as a by-product. The grades of such uranium ores are often in the range 250 4000 ppm or greater. Deep excavations sited in granite would most likely experience radon accumulations sourced from uranium and thorium in the 3-100 ppm range; i.e. a relatively low-level radiation environment. What must be dealt with during site characterization is (1), the possibility that an anomalously high radon source could be encountered (e.g. a portion of the excavation where background uranium and thorium and hence radon emanation increases by a factor of five or ten or more), and (2), the health effects of

long term exposure to such a source particularly if the excavation is a sealed system and the ventilation system is not recirculating air to the atmosphere.

38. It is also pertinent to note that a significant number of scientists feel that far too little is known about the long term health and genetic effects of prolonged exposure to low-level radiation environments. Radon has been highlighted as an important actor in the low level radiation environment and the mechanisms of its mobilization and concentration are still poorly understood in deep geological environments. There has occurred recently a proliferation of meetings, conferences, and studies, dedicated to radon and its concentration in human habitations and dwellings of all sorts as well as its impact on health safety.

Radon Detection and Monitoring Systems

- 39. All systems of radon detection and monitoring are based on the detection of alpha-particles emitted during the decay of radon and its daughters. The methods used to detect and measure alpha particles are (1) the gold leaf electroscope which is now obsolete and of historical interest only; (2), the zinc sulfide scintillator; (3), the ionization chamber; (4), the alpha-track method which is an excellent passive system to determine alpha background and (5), silicon detectors which employ solid state circuitry to record data continuously.
- 40. The alpha-track method is an excellent passive system for the determining the alpha background activity (and anomalies in background) in geological environments and can discriminate between radon and thoron. Alpha sensitive photographic film record the passage of alpha particles along their trajectory as tiny tracks. These tracks can be made visible (microscopically) either photographically or by a method developed by General Electric Co. using cellulose nitrate film which is sensitive to alpha particles but not to light. This method has wide application in the exploration industry, fault and cavity detection studies, earthquake prediction studies, geothermal investigations, and in some environmental monitoring exercises. Alpha particle activity is measured over minimal three-week periods to allow for statistical variations in background caused by atmospheric, wind, and temperature effects to be "meaned-out". The alpha-track method is marketed under the trade name "Track-Etch" by Terradex Corporation (an affiliate of Tech/Ops, Inc.). A similar

system (AlphaCARD) is marketed by AlphaNuclear, a Canadian company who also markets AlphaDOSIMETER and AlphaLOGGER both of which have application in exploration surveys when soil gas radon surveys are desired.

41. The AlphaDOSIMETER system uses solid state silicon detectors, amplifier, and microprocessor to record and store alpha particle activity. Its major application is in underground mines or excavations where personal or environmental measurements are desirable. Exposure is available on a daily basis so that remedial or corrective measures can be taken promptly to minimize personal exposure. Computer storage enables health and radiation personnel, ventilation engineers, etc., to further treat the data statistically; to establish trends and to optimize ventilation and minimize radiation exposure.

The second process of the second process of the second sec

RESEARCH DIRECTIONS

- 42. This study identified several areas of radon behavior research which would benefit site location, characterization, construction and operation of deep underground facilities. The nature of this research is such that several of the topics listed below could be combined as a single research project.
 - <u>a</u>. The geochemical behavior of radon precursors in relation to groundwater geochemistry and physical controls of radon anomalies in the natural environment.
 - b. Mechanisms of radon mobilization and how they vary according to specific geological environments.
 - c. The extent of radon mobilization with depth by atmospheric "pumping" and convection mechanisms.
 - $\underline{\mathbf{d}}$. The extent of radon mobilization by the "sweeping" effect of other gases.
 - Continued support of instrumentation for better detection and monitoring of the natural radiation environment. There is a need for scintillation equipment not subject to contamination and with a low sensitivity to normal light.
- 43. The effect of ionizing radiation produced by radon on the substrates of sophisticated semi-conductors is an open question at this time, (pers. comm. Dr. H. W. Alter, Terradex Corporation). It would be useful to know exactly what information has been gained regarding this aspect of radon decay by such research establishments as Bell Laboratories, The National

Testing Laboratory, International business Machine (IBM), other private sector entities, and the research facilities of the Department of Defense.

44. It is recommended that a research review be conducted to ascertain the state of knowledge and the experimental evidence treating the effect of ionizing radiation produced by radon decay on sophisticated electronic firmwave circuitries, particularly those dedicated to communications and digital signal analysis.

REFERENCES

Department of Geology, Rice University, 1970-1972. "Development of Remote Methods for Obtaining Soil Information and Location of Construction Materials Using Gamma Ray Signatures for Project THEMIS", 5 Vols., Contract No. DACA 39-69-C-0048, US Army Engineer Waterways Experiment Station, Vicksburg, MS.

Doe, B., and M. Newell, 1965, Isotopic Composition of Uranium in Zircon," Am. Mineralogist, Vol 50, p 613-618.

Faul, Henry. 1954. Nuclear Geology, John Wiley and Sons, New York.

Fleisher, R. L., P. B. Price, and R. M. Walker, 1964. "Fission-Track Ages of Zircons," Jour. Geoph. Research, Vol 69, No. 22, p 4885-4888.

. 1965. "Effects of Temperature, Pressure, and Ionization of the Formation and Stability of Fission Tracks in Minerals and Glasses," Jour. Geoph. Research, Vol 70, No. 6, p 1497-1502.

Res. Lab. Reprint 5269. 11 p.

Gableman, John W. 1972. "Radon Emanometry of Starks Salt Dome," US Atomic Energy Commission, RME-4114, 74 p.

Gingrich, James E. and J. C. Fisher, 1976. "Uranium Exploration using the Track Etch Method," IAEA/SM/208-19, Vienna, Austria.

SEAT PARALAGE SELECTION OF THE PROPERTY OF THE PARALAGE SELECTION OF T

LeGrande, Harry E., 1986. "Radon and Radium Emissions from Fractured Crystal-line Rocks--A Conceptual Hydrogeological Model," (Abs), Geological Society of America, Vol 18, No. 3.

Rosholt, J. N., W. R. Shields and E. L. Garner, 1963, "Isotopic Fractionation of Uranium in Sandstone," Science, Vol 169, p 224-226.

Smith, A. Y., P. M. C. Barretto, and S. Pournis, (1976), "Radon Methods in Uranium Exploration," IAEA/SM/208-52, Vienna, Austria.

Thurber, D. L. 1968. "Anomalous U-234/U-238 in Nature," Jour. Geoph. Research, Vol 67, No. 11, p 4518-4520.

BIBLIOGRAPHY

Adams, J. A. S., 1964, Laboratory-Ray Spectrometer for Geochemical Studies. The Natural Radiation Environment, In Adams and Lowder, Editors: The Natural Radiation Environment, Chicago Press, p 485-497.

Adams, J. A. S., Barretto, P. M. C., Clark, R. B. and Duval, J. S., 1971. Radon-222 Emanation and the High Apparent Lead Isotope Ages in Lunar Dust. Nature 231, p 174-175.

Ahrens, L. H., 1955, The Convergent Lead Ages of the Oldest Monazites and Uraninites (Rhodesia, Manitoba, Madagascar and Transvaal), Geoch. Cosm. Acta, Vol 7, p 294 and Vol 8, p 1-15.

Aitken, M. J., Zimmerman, D. W., and Fleming, S. J., 1968. Thermo-Luminescent Dating of Ancient Pottery. Nature, Vol 219, No. 5153, p 442-445.

Amirkhanov, Kh. I., Brandt, S. B., Bartnitskiy, Ye. N., Gasanov, S. A. and Gurvich, J. S., 1958. The Mechanisms of Argon Loss in Micas. Akad. Nauk. SSSR Izv. Ser. Geol. No. 3, p 107-109.

Balacek, K. J., 1966, Radioactivity in Tektites. M.A. Thesis, Rice University.

Barretto, P. M. C., 1971, Radon-222 Emanation from Rocks, Soils and Lunar Dust, M.A. Thesis, Rice University.

Blanchard, R. L., 1969, Radon-222 Daughter Concentrations in Uranium Mine Atmospheres. Nature, Vol 223, p 287-289.

Boltwood, B. B., 1905, The Origin of Radium, Phyl. Mag. and Jour. Sci., Vol IX, Sixth Series, p 603-613.

Boltwood, B. B., 1907, On the Ultimate Disintegration Products of the Radio-active Elements. Part II. The Disintegration Products of Uranium. Am. Jour. Sci., Vol XXIII, No. 134, Fourth Series, p 77-88.

Boltwood, B. B., 1908, Radioactivity of Uranium Minerals. Am. Jour. Sci., Vol XXV, No. 148, Fourth Series, p 269-298.

Brace, W. F., Silver, E., Hadley, K., and Goetze, 1972. Cracks and Pores: A Closer Look. Science, Vol 178, p 162-163.

Catanzaro, E. J., and Hanson, G. N., 1971, U-Pb Ages for Sphene from Early Precambrian Igneous Rocks in Northeastern Minnesota, Northwestern Ontario. Canadian Journal of Earth Sciences, Vol 8, No. 10, p 1319-1324.

Cherdyntsev, V. V., 1971, Uranium-234. (Keter Press, Israel), Chapter III.

Crozier, E. D. and Biles, N., 1966. Measurement of Rn^{220} in the Atmosphere Below 50 cm., Jour. Geophys. Res., Vol 71, p 4735-4741.

Doe, B., and Newell, M., 1965, Isotopic Composition of Uranium in Zircon. Am. Mineralogist, Vol 50, p 613-618.

Doe, B. R. and Pearson, R. D., 1969, U-Th-Pb Chronology of Zircons from the St. Kevin Granite, Northern Sawatch Range, Colorado. Geol. Soc. Am. Bull., 80, p 2495.

Doe, R. R., Lead Isotopes, 1970, (Springer-Verlag).

が、これできた。これでは、これでは、これできたが、これできない。これできない。これできないが、これできない。これできない。これできない。これできない。これできない。これできない。これできない。これできない。これできない。

Druilhet, A., 1966, Diffusion de la radioactivite naturelle de l'air dans les basses couches de l'atmosphere. These doctorat de specialate 405. Faculte des Sciences de Toulouse.

Dyck, W. and Smith, A. Y., 1969. The use of Radon-222 in Surface Waters in Geochemical Prospecting for Uranium. Proc. Intern. Geoch. Expl. Symposium. Quarterly of the Colorado School of Mines, Vol 64, No. 1, p 223-236.

Eckelmann, W. R. and Kulp, J. L., 1956, Uranium Lead Method of Age Determination Part I., Bull. Geol. Soc. Am., Vol 67, p 36-54.

Fireman, E. L., D'Amico, J., Defelice, J., and Spannagel, G., 1972, Radio-activities in Returned Lunar Materials. Proc. Third Lunar Sci. Conf., Vol 2, p 1747-1762.

Fleischer, R. L., Price, P. B. and Walker, R. M., 1964, Fission-Track Ages of Zircons. Jour. Geoph. Res., Vol 69, No. 22, p 4985-4888.

. 1965. Effects of Temperature, Pressure, and Ionization of the Formation and Stability of Fission Tracks in Minerals and Glasses. Jour. Geophy. Res., Vol 70, No. 6, p 1497-1502.

- Fleischer, R. L., Price, P. B. and Walker, R. M. 1966. Tracks of Charged Particles in Solids. General Electric, Res. Lab. Reprint 5269. 11 p.
- Fleischer, R. L., Alter, H. W., Furman, S. C., Price, P. B. and Walker, R. M., 1972, Particle Track Etching. Science, Vol 178, p 255-263.
- Foote, R. S., 1964, Time Variation of Terrestrial Radiation. The Natural Radiation Environment, In Adams and Lowder, Editors: The Natural Radiation Environment, Chicago Press, p 757-766.
- Foyn, E., 1938, Uber einige verhaltnisse in Uranmineralien. Norske vid-Akad., Oslo Skrifter, No. 4.
- Friesen, L. J., 1972, A Theoretical Study of Radon Diffusion in the Lunar Regolith. M.S. Thesis, Rice University. The Moon, Vol 3, p 461-471.
- Gentry, R. V., 1971, Radiohalos: Some Unique Lead Isotope Ratios and Unknown Alpha Radioactivity. Science, Vol 173, p 727-730.
- Giletti, B. J. and Kulp, J. L., 1954, Radon Leakage from Radioactive Minerals. Am. Min., Vol 40, p 481-496.
- Gold, S., Barkhay, H. W., Shllein, B., and Kahn, B., 1964, Measurement of Naturally Occurring Radionuclides in Air The Natural Radiation Environment, In Adams and Lowder, Editors: The Natural Radiation Environment, Chicago Press, p 369-382.
- Goldich, S. S., Nier, A. O., Baadsgaard, H., Hoffman, J. H. and Krueger, H., 1961, The Precambrian Geology and Geochronology of Minnesota. Minnesota Geol. Survey Bull., No. 41, p 193.
- Goldich, S. S., Hedge, C. E., and Stern, T. W., 1970, Age of Morton and Montevideo Gneisses and Related Rocks, Southwestern Minnesota. Geol. Soc. Am. Bull., 81, p 3671-3695.
- Gorenstein, P., Bjorkholm, P., 1972, Observation of Lunar Radon Emanation with the Apollo 15 Alpha Particle Spectrometer. Proc. Third Lunar Sci. Conf., Vol 3, p 2179-2187.
- Gorenstein, P., Bjorkholm, P., 1973, Detection of Radon Emanation from the Crater Aristarchus by the Apollo 15 Alpha Particle Spectrometer, Science, Vol 179, p 792-794.
- Grunenfelder, M. H., 1963, heterogeneity of Accessory Zircons and the Petrographic Significant of their Uranium Decay Age. Schweiz. Mineral. und Petrol. Mitt., No. 43, p 235-257.
- Grunenfelder, M. H., Hofmanner, F., Grogler, N., 1964, Heterogenitat Akzessorischer Zirkone und die Petrographische Deutung Ihrer Uran/Bleizerfallsalter, Schweiz. Mineral. Petrog Mitt. No. 44, p 543-558.
- Guedalia, D., Laurent, J. L., Fontan, J., Blanc, D., Druilhet, A., 1970, A Study of Radon Emanation from Soils, Jour. Geophy. Res., Vol 75, No. 2, p 357-369.
- Hanson, G. N., Catanzaro, E. J., Anderson, D. H., 1971, U-P Ages for Sphene in a Contact Metamorphic Zone. Earth and Planetary Science Letters 12, p 231-237.
- Hanson, G. and Malhotra, R., 1971, K-Ar Ages of Mafic Dikes and Evidence for Low Grade Metamorphism in Northeast Minnesota. Geol. Soc. Am. Bull., Vol 82, p 1107-1113.

Harley, J. H., 1953, Sampling and Measurement of Air-Borne Daughter Products of Radon. Nucleonics, Vol 11, No. 7, p 12-15.

Hayase, I. and Tsutsumi, T., 1958, On the Emanating Power of Powdered Rocks. Memoirs College of Sciences. Kyoto University, Series B., Vol 24, No. 4, p 316-323.

Heymann, D., Yaniv, A., 1971, Distribution of Radon-222 on the Surface of the Moon. Nature Physical Science, Vol 233, No. 37, p 37-39.

Holbrook, G. B. and Weaver, P., 1932, Radon Content of Soil Gas. Physics, Vol 2, No. 5, p 376-385.

Holland, H. D. and Gottfried, D., 1955, The Effect of Nuclear Radiation on the Structure of Zircon. Acta Cryst, 8, p 291-300.

Holmes, A., 1948, The Oldest Known Minerals and Rocks. Edinburg Geol. Soc. Bull., Vol 14 pt 2, p 176-194.

Horne, J. E. T. and Davidson, C. F., 1955, The Age of Mineralization of the Witwatersrand, Bull. Geol. Survey Gt. Brit., No. 10, p 58-73.

Hurley, P. M. and Fairbairn, H. W., 1953, Radiation Damage in Zircon: A Possible Age Method., Geol. Soc. Am. Bull., Vol 64, p 659-673.

Ikebe, Y., 1970, Variation of Radon and Thoron Concentrations in Relation to Wind Speed. Jour. Metereol. Soc. Japan, Vol 48, No. 5, p 461-468.

Ikebe, Y. and Kawano, M., 1970, Dependence of the Effective Attachment Coefficient of Small Ions upon the Size of Condensation Nuclei. Review of Pure and Applied Geophysics, Vol 83, p 120-130.

Ikebe, Y., Shimo, M. and Kawano, M., 1970, Measurements of the Effective Attachment Coefficient Between RaA lons and Condensation Nuclei. Review of Pure and Applied Geophysics, Vol 83, p 131-141.

Ikebe, Y. and Shimo, M., 1972, Estimation of the Vertical Turbulent Diffusivity from Thoron Profiles. Tellus Vol 24, No. 1, p 29-37.

Israel, H. and Israel, G. W., 1965, Measurements of the Thoron Content of the Atmosphere and their Application in Meteorology. Final Technical Report to contract: DA-91-591-EUC-3483.

Israel, H., Horbert, M., and de La Riva, C., 1967, The Thoron Content of the Atmosphere and its Relation to the Exchange Conditions. Final Technical Report to Contract: DA-91-591-EUC-3761.

Israel, H., 1968, Thoron Probleme. Jahrbuch, 1968. Landesamt fur Forschung, des Landes Nordrhein-Westfalen, p. 159-171, Westdeutscher Verlag.

Jaeger, J., 1957, The Temperature in the Neighborhood of a Cooling Intrusive Sheet. Am. Jour. Science, Vol 255, p 306-317.

Kouvo, O., 1958, Radioactive Age of some Finnish Pre-Cambrian Minerals. Comm. Geol. Finlande Bull. No. 182, p 1-70.

Kouvo, O. and Tilton, G. R., 1966, Mineral Ages from the Finnish Pre-Cambrian. Jour. Geol., Vol 74, No. 4, p 421-442.

Kraner, H. W., Schroeder, G. L., and Evans, R. D., 1964, Measurements of the Effects of Atmospheric Variables on Radon-222 Flux and Soil Gas Concentrations, The Natural Radiation Environment, In Adams and Lowder, Editors: The Natural Radiation Environment, Chicago Press, p 191-215.

Kraner, H. W., Schroeder, G. L., Davidson, G., Carpenter, J. W., 1966, Radio-activity of the Lunar Surface. Science, Vol 152, p 1235-1236.

Kulp, J. L., Bate, G. L., Broecker, W. S., 1954, Present Status of the Lead Method of Age Determination, Am. Jour. Sci., Vol 252, p 345-365.

Kuroda, P. K., 1950, On the Isotopic Constitution of Radium (Ra-223/Ra-226) in Uranium Minerals and Recent Problems of Geochronology. Annals N. York Acad. Sciences, Vol 62, p 179-207.

Lambert, G., Grjebine, T., Bristeau, P., Le Roulley, 1973, Excess of Polon-ium 210 at the Surface of Apollo 15 Fines. Abstract Fourth Lunar Sci. Conf., p 452-454.

Leventhal, J. S., 1972, Chronology and Correlation of Young Basalts by Uranium-Thorium-Helium Measurements. Ph.D. thesis, Department of Geosciences, University of Arizona.

Lind, S. C. and Whittemore, C. F., 1914, The Radium: Uranium Ratio in Carnotites. Jour. Am. Chem. Soc., Vol XXXVL, p 2066-2082.

Lindstrom, M., Evans, J. C., Finkel, R. and Arnold, J. R., 1971, Radon Emanation from the Lunar Surface. Earth and Plan. Sci. Let. 11, p 254-256.

Lucas, H. F., 1957, Improved Low-Level Alpha Scintillation Counter for Radon. Review of Scientific Instruments, Vol 28, No. 9, p 680-683.

では、10mmのである。これのなどのなどは、10mmのではないないない。 これのないない 10mmのできない 10mmのできない 10mmのできない 10mmのできないのでは、10mmのできないのでは、10mmのできない。 10mmのできない 10mmのでも 10mmのでをものできない 10mmのできない 10mmのできない 10mmのでものでものでものできない 10mmのでものでものでものでものでものでものでものでものでものでものでものでもの

Lucas, H. F. and Woodward, D. A., 1964, Effect of Long Decay Chains on the Counting Statistics in the Analysis of Radium-224 and Radon-222. Jour. Appl. Physics, Vol 35, No. 2, p 452-456.

Lucas, H. F. and Gabrysh, A. F., 1966, Radon in Coal Mines. Arg. Nat. Lab. Radiological Phys. Division Annual Report ANL-7220, p 58-60.

Megumi, K. and Mamuro, T., 1972, A Method for Measuring Radon and Thoron Exhalation from Ground. Jour. Geophys. Research, Vol 77, No. 17, p 3052-3056.

Moses, H., 1960, The Effect of Meteorological Variables upon the Vertical and Temporal Distributions of Atmospheric Radon, J. Geophys. Res. 65, p 1223-1238.

Moses, H., Lucas, H. F., Jr., and Zerbe, Gunther A., 1063, The Effect of Meteorological Variables upon Radon Concentration Three Feet Above the Ground. APCA Journal, Vol 13, No. 1, p 12-19.

Neuvonen, K. J., 1970, Paleomagnetism of the Dike Systems in Finland v. Remanent Magnetization of the Ava Intrusives. Geol. Soc. Finland Bull. No. 42, p 101-107, The addendum.

Nier, H. O., 1939, The Isotopic Constitution of Radiogenic Leads and the Measurement of Geological Time II, Phys. Rev., Vol 1, p 150-153.

Nycolaysen, L. O., 1957, Solid Diffusion Radioactive Minerals and the Measurement of Absolute Geologic Age, Geochim. Cosmochism. Acta, Vol II, p 41.

Pearson, J. E., 1965, Radon-222 A Study of its Emanation from Soil, Source Strength and Use as Tracer. Final report. University of Illinois.

Pearson, J. E., 1967, Natural Environmental Radioactivity from Rn-222. Public Health Service Publication, No. 999-RH-26.

Pearson, J. E. and Jones, G. E., 1966, Soil Concentrations of Emanating Radium 222 from Soils and Plants. Tellus, Vol 18, No. 2-3, p 655-662.

Pellas, P., 1965, Etude Sur la Recristallisation Thermique des Zircons Metamictes. Serie C. Sciences de la Terre, Tome XII Fasc. 5, p 227-253.

Pidgeon, R. T., Oneil, J. R. and Silver, L. T., 1966, Uranium and Lead Stability in a Metamict Zircon under Experimental Hydrothermal Conditions. Science, Vol 154, p 1538-1540.

Pohl-Ruling and Pohl, E., The Radon-222 Concentration in the Atmospheres of Mines as a Function of Barometric Pressure. Health Physics, Vol 16, p 579-584.

Perry, E., Jr., and Bonnichsen, 1966, Quartz and Magnetite: $0^{18}/0^{16}$ Fractionation Profile from Metamorphosed Biwabik Iron Formation. Science, Vol 153, p 528-529.

Rankama, K., and Sahama, TH G., Geochemistry, The University of Chicago Press, 1968 Edition.

Rankin, D. W., Stern, T. W., Reed, J. C., and Newell, M. F., 1969, Zircon Ages of Felsic Volcanic Rocks in the Upper Pre-Cambrian of the Blue Ridge, Appalachian Mountains. Science, Vol 166, p 741-744.

Rosholt, J. N., Shields, W. R. and Garner, E. L., 1963, Isotopic Fractionation of Uranium in Sandstone, Science, Vol 169, p 224-226.

Schroeder, G. L., Kraner, H. W. and Evans, R. D., 1965, Diffusion of Radon in Several Naturally Occurring Soil Types. Jour. Geophys. Res., Vol 70, No. 2, p 471-474.

Silver, L. T., 1963(a), The Use of Cogenetic Uranium Lead Isotope Systems in Zircons in Geochronology, In Radioactive Dating, Intern, Atomic E. Agency Symposium, Athens, Proc. p 279-287.

Silver, L. T., 1963(b), The Relation Between Radioactivity and Discordance in Zircons. In Nuclear Geophysics, Nat. Acad. Sciences - NRC publ. 1075, p 34-42.

Silver, L. T., 1970, Uranium-Thorium-Lead Isotopes in some Tranquility Base Samples and their Implications on the Lunar History. Proc. Apollo 1! Lunar Sci. Conf., Vol 2, p 1533-1574.

Silver, L. T., 1972(a) U-Pb-Th Abundances and Isotopic Characteristics in some Apollo 14 Rocks, Soils and an Apollo 15 Soil. Abstract. Third Lunar Science Conference, p 620.

Silver, L. T., 1972(b) Lead Volatilization and Volatile Transfer Processes on the Moon. Abstract 3rd Lunar Science Conf., p 617-619.

Silver, L. T. and Deutsch, S., 1963(b) Uranium-Lead Isotopic Variations in Zircons, A Case Study. J. Geol., Vol 71, No. 6, p 721-758.

Steiger $^{R.2H_8}$, Wasserburg, G. J., 1966, Systematics in the Pb 208 -Th $^{-232}$ -U and Pb $^{-238}$ -U Systems. J. Geophys. Res., Vol 71, No. 24, p 6065-6090.

Stern, T. W., Goldich, S. S., Newell, M. R., 1966, Effects of Weathering on the U-Pb Ages of Zircon from the Morton Gneiss, Minnesota, Earth, Plan. Sci. Let., 1, p 369-371.

Stieff, L. R. and Stern, W., 1955, Interpretation of the Discordand Age Sequence of Uranium Ores., US Geol. Survey Prof. Paper 300, p 549-555.

Stoenner, R. W., Lyman, W. and Davis, Jr., R., 1971, Radioactive Rare Gases and Tritium in Lunar Rocks and in the Sample Return Container, Proc. Sec. Lunar Sci. Conf., Vol 2, p 1813-1823.

Stoenner, R. W., Lindistrom, R. M., Lyman, W. and Davis Jr., R., 1972, Argon, Radon and Tritium Radioactivities in the Sample Return Container and the Lunar Surface. Proc. Third Lunar Sci, Conf., Vol 2, p 1703-1717.

Tanner, Allan B., 1964, Radon Migration in the Ground, A Review, in <u>The Natural Radiation Environment</u>. In Adams and Lowder, Editors: The Natural Radiation Environment Chicago Press, p 161-190.

Tanner, Allan B., 1964, Physical and Chemical Controls on Distribution of Radium-226 and Radon-222 in Ground Water near Great Salt Lake, Utah, in The Natural Radiation Environment. In Adams and Lowder, Editors: The Natural Radiation Environment, Chicago Press, p 253-276.

Tatsumoto, M., 1970, Age of the Moon: An Isotopic Study of U-Th-Pb Systematics of Apollo II Lunar Samples II. Proc. Apollo II Lunar Sci. Conf., Vol 2, p 1595-1612.

Tatsumoto, M., Knight, R. J. and Doe, B. R., 1971, U-Th-Pb Systematics of the Apollo 12 Lunar Samples, Proc. Second Lunar Science Conference, p 1521-1546.

Tatsumoto, M., Hedge, C. E., Doe, B. R., Unruth, D., 1972, U-Th-Pb and Rb-Sr Measurements on some Apollo 14 Lunar Samples. Abstract Third Lunar Sci. Conf., p 655.

Tilton, G. R., 1960, Volume Diffusion as a Mechanism for Discordant Lead Ages, Jour. Geoph. Res., Vol 65, No. 9, p 2933-2945.

Tilton, G. R. and Grunenfelder, M. H., 1968, Sphene: Uranium Lead Ages, Science, Vol 159, p 1458-1461.

Thurber, D. L., 1962, Anomalous U^{234}/U^{238} in Nature, Jour. Geoph. Research, Vol 67, No. 11, p 4518-4520.

Turkevich, A. L., Patterson, J. H., Franzbrote, E. J., Sowinski, K. P., Economou, T. E., 1970, Alpha Radioactivity of the Lunar Surface at the Landing Sites of Surveyors 5, 6, and 7. Science, Vol 167, p 1722-1724.

Vaz, J. E. and Senftle, F. D., 1971, Thermoluminescence Study of the Natural Radiation Damage in Zircon, Jour. of Geoph. Res., Vol 76, No. 8. p 2038-2050.

Wasserburg, G. J., 1963, Diffusion Processes in Lead-Uranium Systems, Jour. Geoph. Res., Vol 68, No. 16, p 4823-4846.

Wetheril, G. W., Kouvo, O., Tilton, G. R., and Gast, P. W., 1962, Age Measurements on Rocks from the Finnish Pre-Cambrian, Jour. Geol., Vol 70, No. 1, p 74-88.

Wetheril, G. W., 1956, Discordant Uranium Lead Ages, I Trans. Amer. Geoph. Union, Vol 37, No. 3, p 320-326.

Wetheril, G. W., 1963, Discordant Uranium, Lead Ages, Jour. Geoph. Res., Vol 68, No. 10, p 2957-2965.

Wilson, J. T., Farquhar, R. M., Gretener, P., Russel, R. D., and Shillibeer, H. A., 1954, Estimate Age for some African Minerals, Nature, Vol 174, p 1006-1007.

Wilkening, M. H., and Hand, J. E., 1960, Radon Flux at the Earth-Air Interface, Jour. Geophys. Res., Vol 65, p 3367-3370.

Yaniv, A. and Heymann, D., 1972, Radon Emanation from Apollo 11, 12 and 14 Fines, Abstract Third Lunar Science Conference, p 718-720.

Yeh, R. S., Allen, J. A. V., 1969, Alpha-Particle Emissivity of the Moon: An Observed Upper Limit, Science, Vol 166, p 370-372.

APPENDIX A: NOTATION

eU: Equivalent uranium content or grade as measured by calibrated gamma ray spectrometer, scintillometer, or similar detection system. Usually expressed in parts per million (ppm).

Ci: Curie; the unit of activity in the field of radiation dosimetry.

One curie equals 3.7 x 1010 disintegrations per second. (The activity of one gram of radon-226 is slightly less than 1 curie.

pCi: Pico curie; equal to 10-12 curies.

222-Rn Activity and Emanation Rate:

Barretto (1971) details the laboratory, alpha-counting, mathematical, and data processing methods and procedures used to determine both 222-Radon activity and emanation rate.

The amount of alpha particles produced by a radon source material over a set (measurable) period of time, is taken as an indicator of radon activity and is given here in units of picocuries/hour/ gram \times 10^{-4} .

The emanation rate is derived from a differential equation that describes the rate of change of the number of radon and daughter isotope atoms as a function of time. Emanation rates shown herein are given in units of atoms/hour/gram.

Escape-to-Production Ratio (%):

The escape-to-production ratio (EPR) is given as a percentage of radon activity and calculated from the equation:

EPR = 100 - [(E - a)/A],

where ${\bf E}$ is the emanation rate and ${\bf A}$ the experimentally derived radon activity.

US Standard Sieve Mesh Numbers:

The mesh numbers in the Tables and Figures from Barretto (1971) refer to the US Standard Sieve Mesh Numbers which correspond to the following mean grain sizes after sample crushing and sieving:

60 Mesh = 0.250 mm (250 microns) 100 Mesh = 0.149 mm (149 microns) 115 Mesh = 0.129 mm (129 microns) 200 Mesh = 0.074 mm (74 microns)

H_() DATE FILMED APRIL 1988 D/1/C